



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(54) Title: PROCESS AND APPARATUS FOR MELAMINE MANUFACTURE</b>		
<b>(57) Abstract</b>  <p>High pressure melamine manufacturing process starting from urea with high yields and conversion rates characterized in that it comprises the following steps: a) urea is fed to a reactor essentially containing molten melamine, which reactor operates in continuous and is kept at a temperature in the range of 360 and 420 °C under a pressure higher than <math>7 \times 10^3</math> KPa and preferably <math>8 \times 10^3</math> to <math>9 \times 10^3</math> KPa, while a vigorous mixing is provided by evolving gases; b) liquid reaction product is recovered containing 85 to 95 %, preferably 88 to 93 % melamine and a gaseous phase is removed containing essentially CO<sub>2</sub> and NH<sub>3</sub>; c) liquid phase collected in b) is continuously fed, together with fresh NH<sub>3</sub> to a tubular reactor, in which the essentially whole volume is occupied by the liquid phase (plug flow reactor) without any mixing of the reaction product with reactant nor the intermediate products (no "back mixing"), kept at a temperature of 360 to 450 °C and under a pressure higher than <math>7 \times 10^3</math> KPa for a residence time sufficient to complete the reaction; d) melamine with a high purity level is collected from the outlet of the tubular reactor.</p>		

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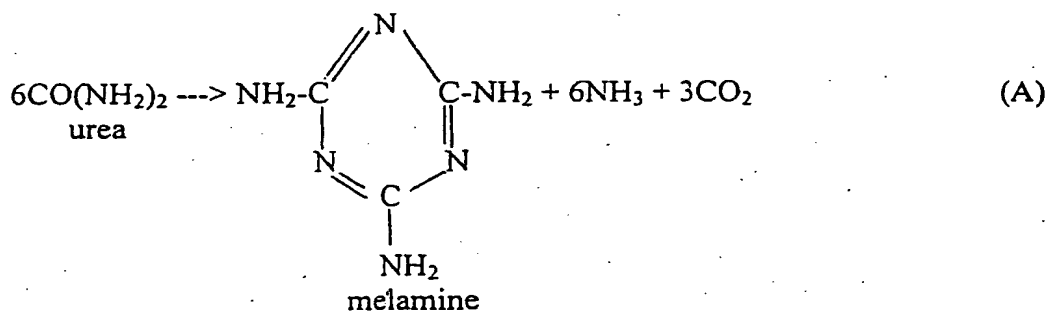
Description"PROCESS AND APPARATUS FOR MELAMINE MANUFACTURE"Technical field

This invention concerns a process for high yield manufacture of high purity melamine  
 5 and the apparatus to carry out the process.

More particularly this invention is directed to melamine preparation starting from urea  
 by means of a so-called high pressure process.

Background art

Melamine is presently manufactured from urea according to the following simplified  
 10 reaction scheme:

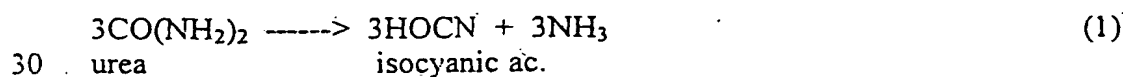


20 The reaction is highly endothermic, the reaction heat at a temperature of 360 to 420°C being approximately 93,000 Kcalories per Kmole of melamine.

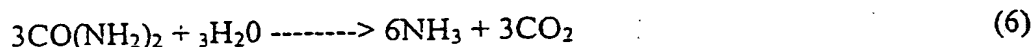
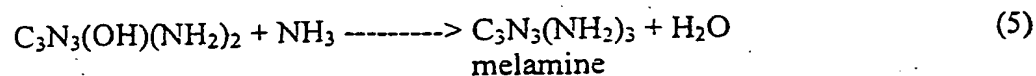
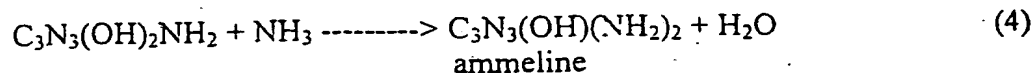
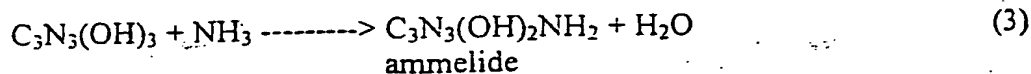
The process can be carried out at low pressure in presence of catalysts or at high pressure without any catalyst.

25 Both low pressure and high pressure melamine manufacturing processes starting from urea are believed to proceed through a series of intermediate reactions leading to, respectively, isocyanic acid, cyanuric acid, ammelide, ammeline and finally melamine.

It seems that the following reactions are involved:



cyanuric ac.



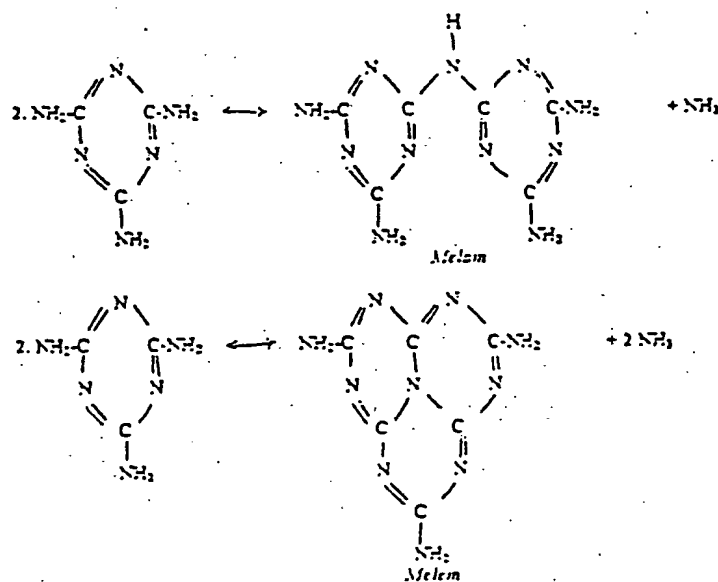
By summing up equations 1 to 6 the overall reaction equation (A) is obtained.

Some of these intermediate products, namely ammeline and ammelide, hereinafter collectively referred to as OAT (OxyAminoTriazine), have been detected in the reaction products.

Moreover melamine, obtained as above, reacts with itself under reaction temperature and pressure yielding so-called polycondensates and a release of ammonia.

Polycondensates, beyond being impurities reducing melamine purity degree decrease also overall reaction yields.

Polycondensates result from amine group (-NH<sub>2</sub>) ammonolysis of the melamine molecule and are formed e.g. according to the following equations:



The above reactions are promoted by a low or nonexistent ammonia partial pressure and the long residence time of melamine in the liquid phase ( $\geq 355^{\circ}\text{C}$ ). In the melamine synthesis conditions polycondensates are obtained in a low amount, however such an amount is not negligible in connection with the final product purity. Anyhow an almost  
5 complete polycondensate to melamine regression is achieved by increasing ammonia partial pressure. In the nowadays melamine synthesis processes, polycondensates conversion to melamine takes place in the melamine purification section wherein, inter alia, an ammonia treatment of the reaction product is provided for.

In the high pressure process, molten urea at a temperature of 140 to 150°C is fed to a  
10 reactor, kept at a temperature of 360 to 420°C by means of suitable heating devices. In this reactor molten urea mixes with melamine and remains under stirring actions of the evolving reaction gases for a determined period of time. Raw melamine product is subject to a purification treatment, for instance by dissolving it in water and subsequent recrystallization to eliminate unreacted urea and remove reaction by-products essentially  
15 consisting of reaction gaseous products (ammonia and carbon dioxide), liquid reaction products essentially comprising OAT (mainly ammeline) and polycondensates.

In the industrial process embodiments, reaction is carried out in continuous, typically in a single reactor consisting of a cylindrical vessel (tank reactor) wherein the reactants are kept under vigorous mixing by the generation and evolution of reaction gaseous  
20 products. Reaction heat is supplied to reactants through suitable heat exchange tubes in which molten salts circulate at a temperature higher than the reaction one.

Inside the reactor each chemical species concentration exhibits a constant value almost in any point of the liquid reaction mixture. Molten urea, continuously fed to the reaction zone, immediately mixes with circulating reaction mixture. The reaction product is  
25 continuously removed and it has the same concentration as the reaction mixture in the

reactor. In said reaction arrangement, the higher is the desired conversion rate, and the lower is melamine production rate. Therefore large reaction volumes are required resulting in a very expensive operation in that the reactor has to be resistant to the highly corrosive action of the reactants and reaction products under very severe temperature and pressure conditions. As a consequence the costs of the material of the reactor and its working are extremely high.

Even though the reactor had a reactor volume sufficient to achieve approximately a 100% conversion ratio, by remarkably increasing in such a way the reactor costs, it would not be possible to manufacture melamine at a purity degree as required by the market. As a matter of fact, on one side, even optimizing the mixing of reaction mixture, it is not possible to prevent part of the reactants (urea) from coming out of the reactor before the necessary residence time is elapsed to enable its complete dissolution into the liquid mass and its complete conversion to melamine. The smaller is the reaction volume the more is the content of unreacted components which is present in the reaction mixture. Moreover unreacted component content increases, depending on the departing of the reaction mixture from the ideal mixing conditions. On the other side the residence time distribution in the reaction is such as that roughly one half of the reactant mixture remains inside the reactor for a period of time longer than the average residence time, i.e. the ratio between the reactor volume and reactant volume flow rate. Since the reaction mixture practically consists of melamine only, it is subject for a long period of time to ammonolysis reaction resulting in an increased amount of polycondensates.

Therefore single reactor melamine manufacturing processes yields low purity degree melamine (lower than 97 - 98%) suitable for marginal uses only, unless the reaction product is submitted to purification treatments affecting the process overall economy, to reach high purity melamine (higher than 99.5%).

Multiple reaction section melamine synthesis processes have been proposed which allow to increase the melamine purity. An example of a two-step melamine synthesis process has been disclosed in US-A-3,116,294. However since the second reactor employed in the second step is analogous to the first, i.e. both are tank reactors, same  
5 drawbacks are experienced, even of a less importance, as in the single reactor process.

#### Disclosure of invention

It would be highly desirable to have available a high yield and conversion, high purity melamine manufacturing process which does not require expensive apparatuses.

It is therefore an object of the present invention a high pressure melamine  
10 manufacturing process starting from urea which process allows to obtain with high conversions a high purity product that may be employed as such, or after a simple purification, for most of the conventional uses. A further object of the present invention is an apparatus that enables to carry out the above process.

In particular the present invention is based upon a reaction system carried out in two or  
15 more consequent reaction steps wherein, from the first through the latter step, the reaction parameters as reaction overall pressure and temperature, and gaseous product partial pressures are progressively changed; residence times being thoroughly controlled especially in the last reaction steps in order to accomplish the complete reaction and the relevant disappearance of by-products with an increased yield.

20 More particularly the high-pressure melamine manufacturing process starting from urea according to present invention comprises the following steps:

a) urea is fed to a reactor essentially containing molten melamine, which reactor operates in continuous and is kept at a temperature in the range of 360 an 420°C under a pressure higher than  $7 \times 10^3$  kPa and preferably  $8 \times 10^3$  to  $9 \times 10^3$  kPa, while a vigorous  
25 mixing is provided by the evolving gases;

b) liquid reaction product is recovered containing 85 to 95%, preferably 88 to 93% melamine and a gaseous phase is removed containing essentially  $\text{CO}_2$  and  $\text{NH}_3$ ;

c) liquid phase collected in b) is continuously fed, together with fresh  $\text{NH}_3$  to a tubular reactor, in which the essentially whole volume is occupied by the liquid phase (plug  
5 flow reactor) without any mixing of the reaction product with reactants nor the intermediate products (no "back mixing"), kept at a temperature of 360 to 450°C and under a pressure higher than  $7 \times 10^3 \text{ kPa}$  for a residence time sufficient to complete the reaction;

d) melamine with purity level higher than 99.5, not including dissolved gaseous phase,  
10 is collected from the outlet of the tubular reactor.

Liquid phase entering the tubular reactor according to step c) above, passes through the entire reactor length within a precisely defined period of time corresponding to residence time defined by the ratio between tubular reactor length and liquid reacting mixture linear velocity through the reactor itself.

15 The process according to the present invention allows to obtain a high purity melamine with high reaction yield by using a standard type tank reactor where at least one tubular reactor, as above defined, is connected downstream of the standard reactor.

The process according to the present invention can be applied to existing melamine manufacturing plants to obtain higher yields and purity level of the product.

20 It is apparent that the process of the invention may be applied to new melamine synthesis plants. In such cases the process of the invention allows to obtain melamine with a higher yields and purity degree using a smaller size tank reactor which is the more expensive equipment of the overall unit.

The amount of fresh ammonia to be fed to the tubular reactor together with the reaction  
25 liquid mixture from step b) is higher than the sum of the amount corresponding to liquid



mixture saturation plus the stoichiometric amount necessary to convert all OAT and all condensates to melamine. The amount of ammonia will be such as to ensure substantial excess ammonia within the liquid phase.

According to another embodiment of the present invention, the second step of the reaction is divided in two sections. At the end of the first section, gaseous phase, essentially comprising ammonia, carbon dioxide and melamine vapor traces, is removed and molten melamine is fed, together with fresh ammonia and after removal of dissolved carbon dioxide, to the second reaction section comprising a tubular reactor similar to the first section. In this second reaction section the pressure within the tubular reactor is higher than the pressure both of the first reactor (tank reactor) and the first section of the second reaction step.

A further embodiment of the high yield melamine manufacturing process according to the invention may comprise the addition of a third section to the second step of the reaction similar to the second section. However said addition is generally not necessary in that the process according to the invention wherein the second reaction step comprises one or two reaction sections can obtain a very high purity. The configuration with three or more sections in the second reaction step could be useful only in case a very close to 100% melamine purity is sought.

Gaseous phase removed from the reaction product both in the final and intermediate stages may be sent to urea synthesis, after gaseous melamine recovery; alternatively a portion of the ammonia present in said gaseous phase may be separated and used in the process.

Tubular reactor employed in the second step of the process according to the present invention shall have geometrical parameters to ensure a Reynolds number higher than 5,000, preferably higher than 10,000.

Heating means of the step a) reactor is quite conventional and it may consist of molten salt heating coils arranged inside the reactor. Step c)-tubular reactor can be heated by a molten salt bath.

Temperature inside "plug flow" tubular reactor may be the same as the one of the tank reactor, preferably is higher than that to shorten reactant residence times and ensure reaction completeness.

Melamine recovered at the outlet of tubular reactor exhibits purity higher than 99.5%, therefore it may be directly used after cooling and stripping out of dissolved gaseous phase without any additional treatments.

#### 10 Detailed description of the invention and drawings

The process according to the invention will now be described with reference to the accompanying drawing consisting of Figure 1 and 2 as well as the following experimental examples. Examples and drawings are intended to better explain how to practice the invention and the advantages thereof, but they have not to be interpreted as  
15 limiting its scope.

#### EXAMPLE 1

Reference is made to figure 1 wherein a reaction scheme for melamine manufacture from urea is illustrated consisting of a first step reactor 3 combined with a second step reactor 4.

20 First step reactor 3 is kept under operating pressure by means of a pressure regulator 31 which, acting on the control valve 32, ensures a controlled discharge of the reaction gas products through line 33. Said gases consist of ammonia and carbon dioxide in an approximate ratio of two moles of ammonia per mole of carbon dioxide. Said gases are saturated of melamine vapors. First reaction step is continuously fed, through line 11,  
25 with molten urea, which has been compressed up to reaction pressure by means of pump

1. Through line 21 and pump 2, liquid ammonia is also sent in line 11, in order to keep constant the urea feeding to reactor, avoiding therefore any possible generation of plugging as a result of urea decomposition due to reactor high temperature.

The correct temperature value in the first reaction step is ensured by molten salt circulation inside exchange tubes 37 fully plunged into the liquid reaction mixture. In this example reaction temperature is kept at 385°C. Inside the reactor molten reaction mixture is subject to fast movement around central conveyor tube 38 because of the evolution of ammonia and carbon dioxide gases during the reaction. Said gases are relatively poorly soluble in the liquid mass and therefore they go up along the reactor within the annular space between conveyor tube and the wall of the reactor itself where molten salt heating tubes 37 are located. Said gases, after having reached the reaction liquid mixture upper surface, evolve and are conveyed outside the reactor through line 33 and valve 32 whose opening is controlled by pressure regulator 31 to keep the reactor under programmed operating pressure.

A liquid sensor level 34, placed inside the reactor, allows maintaining constant the reactor liquid level by acting upon valve 35 to discharge through line 36 an amount of liquid corresponding to the reactant volume fed to reactor.

Outlet line 36 is placed in a portion of the reactor beneath the liquid level, therefore it may discharge only a liquid phase essentially consisting of melamine containing a certain amount of unreacted urea, polycondensates, OAT, beyond a very little amount of dissolved ammonia and carbon dioxide.

Line 36 is connected to the second reaction step consisting of a tubular reactor fully plunged into a molten salt bath 41, ensuring a homogeneous reaction temperature all along the reactor.

Line 22 runs into line 36 downstream of valve 35. Line 22, by means of pump 2, ensures

an ammonia inlet into reactor 4 to maintain a constant ammonia saturation of reactor liquid volume, in spite of ammonia consumption because of the reaction of the latter with ploycondensates and OAT.

Ammonia coming from pump 2 to feed both reaction steps through line 21 and 22, is  
5 vaporized and superheated up to the reaction temperature by suitable heat exchange means not illustrated in the drawing.

Reaction mixture going through second step of reaction 4 resides inside the reactor, under defined temperature and pressure conditions, for a period of time depending on the tubular reactor length and the liquid linear velocity.

10 At the outlet of reactor 4, reaction liquid mixture, through line 42, enters the gas/liquid separator 5 where liquid melamine is separated from gases, essentially comprising ammonia containing small a amount of carbon dioxide; said gases are injected, through line 53 into line 33. Valve 52, controlled by pressure regulator 51, maintain separator 5 under a pressure very close to the one of first reaction step 3. The pressure in separator 5  
15 is however lower than the pressure of the first reaction step 3 to allow the liquid mass, coming from line 36 and passing through the second reaction step 4, to reach gas/liquid separator 5 overcoming the friction losses of the second reaction step 4, lines 36 and 42 and valve 35.

Liquid melamine is collected from separator 5 through line 56 by means of control  
20 valve 55, which is acted by level sensor 54, allows to keep a constant level in separator 5.

8,230 Kg/h of urea and 410 Kg/h of ammonia are fed to first step reactor as illustrated above.

Reaction pressure is kept to a fixed value of  $8 \times 10^3$  kPa by discharging, under pressure  
25 control, about 5,700 Kg/h off-gas consisting of 46% ammonia by weight, 50% carbon

dioxide by weight and about 4% melamine vapor by weight. Melamine vapor are totally recovered and injected again into the reaction zone by means of a conventional off-gas cooling unit and melamine adsorption using the urea stream charged to reactor; said melamine recovery unit is not illustrated in the drawing. Recovered melamine, in an amount of about 220 Kg/h, returns to reactor through line 11 and therefore any loss of yield is avoided.

Reaction temperature is kept at an average value of 385°C by the circulation of molten salts fed to the reactor at a flow rate of 430m<sup>3</sup>/h at a temperature of 420°C. Outlet temperature of the same is 410°C due to endothermic nature of the reaction.

At the exit of first reaction step (line 36) the following products are obtained:

	melamine	2,709 kg/h
	unreacted urea	75 kg/h
	OAT	110 kg/h
	polycondensates	60 kg/h
15	dissolved ammonia	40 kg/h
	dissolved carbon dioxide	25 kg/h

Melamine to urea conversion according to the overall reaction stoichiometry (see equation 7) is 94% and melamine purity degree excluding gaseous products (ammonia and carbon dioxide) is 91.7%.

150 kg/h of ammonia is added to the liquid mixture coming out of first reaction step before entering the second reaction step.

Second reaction step consists of a tube 234 m long with an internal diameter of 5 cm. Fluid linear velocity inside the tube is 35 cm/sec., Reynolds number being 17,700.

Thermostatic vessel is kept at 420°C using the same molten salts as in first step being practically negligible the amount of heat associated to chemical reactions taking place in

second step reactor. Pressure is fixed to  $7.5 \times 10^3$  kPa in order to compensate the pressure drop due to valve 35. In the above conditions 2,880 kg/h of degassed melamine are obtained with a purity of 99.64% (excluding gaseous products).

The overall reaction yield is 99.62%.

## 5 EXAMPLE 2

Figure 2 illustrates a melamine reaction unit starting from urea equipped with a second reaction step comprising two sections.

Section 2/a is operated at the same temperature and pressure conditions as the second reaction step of Example 1, while section 2/b is operated at a higher pressure ranging  
10 between  $8 \times 10^3$  kPa and  $30 \times 10^3$  kPa and includes the removal of carbon dioxide from the reaction mixture coming out of section 2/a. In this Example the pressure is  $20 \times 10^3$  kPa.

First step reactor is kept under the same conditions as Example 1:

temperature	385°C
pressure	$8 \times 10^3$ kPa
15 urea flow rate (pump 1, line 11)	8230 kg/h
ammonia linear velocity (pump 2, line 21)	410 kg/h

From the outlet of said first reaction step a liquid phase is obtained having the following composition:

20 melamine	2714 kg/h
unreacted urea	75 kg/h
OAT	110 kg/h
polycondensates	60 kg/h
dissolved ammonia	40 kg/h
dissolved carbon dioxide	25 kg/h

25 The above liquid phase is fed, through line 36, to section 2/a of the second reaction step

together with 75kg/h ammonia coming from pump 2 through line 22.

The outcoming stream from section 2/a of the second reaction step (line 42) contains 71kg/h CO<sub>2</sub> partly, namely 25kg/h, deriving from liquid reaction mixture entering reactor 4, and partially (46 kg/h) as a result of conversion to melamine of urea and OAT coming out of the first reaction step.

Pressure and temperature of reactor 4 is, as in Example 1,  $7.5 \times 10^3$  kPa and 420°C respectively. Reactor 4 consists of a tube, 234 meter long with an internal diameter of 5cm, wherein the reaction mixture pass with a linear velocity of 35 cm/sec, Reynolds number being 17,700. Reactor 4 effluent contains raw melamine with a purity degree better than 99%, besides ammonia that is partly in the gaseous state and partly dissolved as well as carbon dioxide, 80% of which is present as a gas phase with ammonia. This effluent, through line 42, enters stripping tower 6, where, after gas phase removal including most of carbon dioxide, liquid phase is subject to a dissolved carbon dioxide stripping by scrubbing said liquid phase with 200kg/h gaseous ammonia at 420°C, coming from pump 2 and line 23.

Pressure is maintained constant at the value of  $7.5 \times 10^3$  kPa in the tower by means of the pressure regulator 61 which, acting on valve 62, discharges gases into off-gas duct through lines 52 and 63.

Liquid phase coming out of tower 6 through line 65 consists of a 2935 kg/h raw melamine stream 99% pure, containing 50 kg/h dissolved ammonia. Effluent from tower 6 is practically free from carbon dioxide.

Tower 6 liquid level is kept constant by means of level control 64 acting on flow rate of the extraction and compression pump 7, which is connected with tower 6 through line 65 on one side and section 2/b tubular reactor 9 through line 66 on the other side. Pump 7 is designed to rise the pressure of reactor 9 to  $20 \times 10^3$  kPa to allow a fast conversion

into melamine of polycondensates which have not been converted in reactor 4. For this purpose, 250 kg/h of superheated gaseous ammonia coming from line 66 are injected into reactor 9 through line 81 by means of a second ammonia pump 8 before the introduction of raw melamine. Ammonia vaporization and superheating devices in lines

5 21 to 23 and 81 are not shown in the drawing to simplify the overall reaction scheme.

Reactor 9 is kept at 420°C by a molten salt bath 91. Reactor consists of a 8 cm internal diameter, 92 meter long tube, where liquid phase pass through at a 14 cm/sec linear velocity corresponding to a Reynolds number of 11,000.

High purity melamine comes out of reactor 9 through line 92, then it passes to gas/liquid  
10 separator 5 where gaseous ammonia is sent to off-gas duct 33 through line 53, under the pressure control provided by the system pressure regulator 51 and valve 52.

Through line 56, under the level control provided by the system level sensor 54 and valve 55, liquid mixture containing 2880 kg/h of pure melamine (>99,9%) is recovered together with 150-160 kg/h dissolved ammonia.

15 Reaction yield, referred to overall equation (7), is practically 100%.



## CLAIMS

1. High-pressure melamine manufacturing process starting from urea with high yields and conversion rates characterized in that it comprises the following steps:

a) urea is fed to a reactor essentially containing molten melamine, which reactor operates in continuous and is kept at a temperature in the range of 360 an 420°C under a pressure higher than  $7 \times 10^3$  kPa and preferably  $8 \times 10^3$  to  $9 \times 10^3$  kPa, while a vigorous mixing is provided by evolving gases;

b) liquid reaction product is recovered containing 85 to 95%, preferably 88 to 93% melamine and a gaseous phase is removed containing essentially  $\text{CO}_2$  and  $\text{NH}_3$ ;

c) liquid phase collected in b) is continuously fed, together with fresh  $\text{NH}_3$  to a tubular reactor, in which the essentially whole volume is occupied by the liquid phase (plug flow reactor) without any mixing of the reaction product with reactants nor the intermediate products (no "back mixing"), kept at a temperature of 360 to 450°C and under a pressure higher than  $7 \times 10^3$  kPa for a residence time sufficient to complete the reaction;

d) melamine with a high purity level is collected from the outlet of the tubular reactor.

2. High pressure melamine manufacturing process starting from urea according to claim 1, characterized in that the amount of fresh ammonia to be fed to the tubular reactor together with the reaction liquid mixture from step b) is higher than the sum of the amount corresponding to liquid mixture saturation plus the stoichiometric amount necessary to convert all OAT and all polycondensates to melamine, the amount of ammonia being such as to ensure a substantial excess ammonia within the liquid phase.

3. High pressure melamine manufacturing process starting from urea according to claims 1 or 2, characterized in that the second reaction step is divided in two sections wherein at the end of the first section, gaseous phase essentially comprising ammonia,

carbon dioxide and melamine vapor traces, is removed and molten melamine is fed, together with fresh ammonia and after removal of dissolved carbon dioxide, to the second reaction section comprising a tubular reactor similar to the first section, the pressure within the tubular reactor of the second reaction section being higher than the pressure both of the first reactor (tank reactor) and the first section of the second reaction step.

4. High-pressure melamine manufacturing process starting from urea according to preceding claim, characterized in that the pressure in the second section of the second reaction step is comprised between  $8 \times 10^3$  and  $30 \times 10^3$  kPa.

5. High-pressure melamine manufacturing process starting from urea according to claims 3 and 4, characterized in that a third section, similar to the second one, is added to the second reaction step.

6. High pressure melamine manufacturing process starting from urea according to the preceding claims, characterized in that the tubular reactor employed in the second step or in the second step sections of the process has geometrical parameters to ensure a Reynolds number higher than 5,000.

7. High-pressure melamine manufacturing process starting from urea according to the preceding claims, characterized in that the heating of tubular reactor of the second step or the sections of second step is obtained by immersion of the reactor in a molten salt bath.

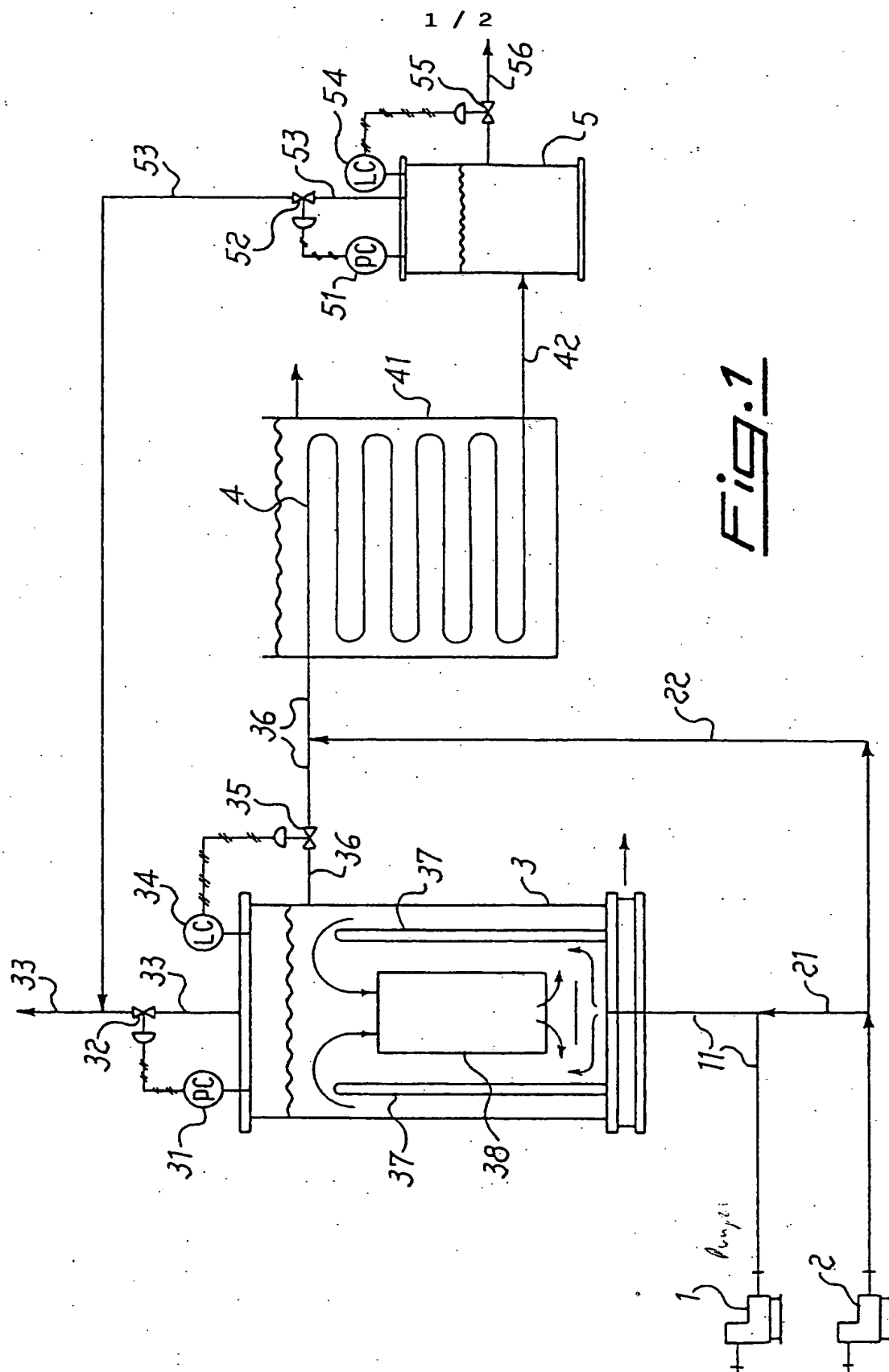
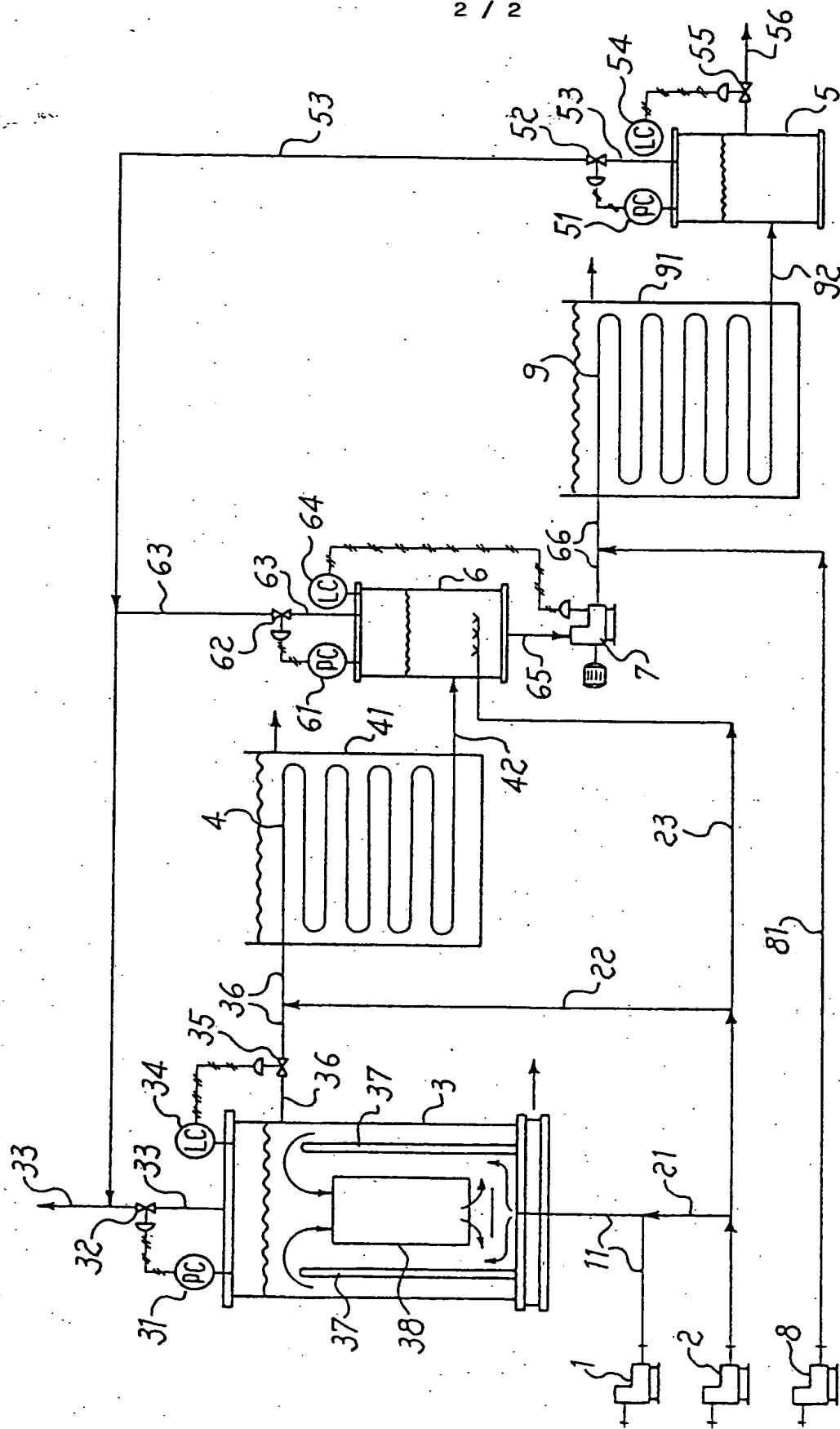


Fig. 1

Fig. 2



# INTERNATIONAL SEARCH REPORT

International Application No

PCT/IT 98/00161

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C07D251/60

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C07D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 3 116 294 A (G. MARULLO ET AL) 31 December 1963 cited in the application * complete document *	1
A	US 5 514 797 A (DAVID BEST) 7 May 1996 * complete document *	1

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

### \* Special categories of cited documents :

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Date of the actual completion of the international search

30 September 1998

Date of mailing of the international search report

08/10/1998

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# INTERNATIONAL SEARCH REPORT

information on patent family members

International Application No

PCT/IT 98/00161

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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